Annihilation of Slow Positrons in Nitric Oxide, Argon, Methane and Deuterated Methane

by

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Abstract

Measurements of positron annihilation rates in nitric oxide,argon, methane and deuterated methane have been made at different pressures of these gases.

In case of nitric oxide the plot of annihilation rate against pressure has been found to be a parabola. An attempt has been made to explain the behaviour of positrons in this gas. The values of "a" and "b" in the formula $\lambda = ap + bp^2$ have been determined as 9.3×10^{-3} μsec^{-1}/mms Hg and 5×10^{-6} μsec^{-1}/(mms Hg)^2 respectively.

The annihilation rate of positrons measured in argon shows the usual linear dependence with pressure. An increase in the pressure coefficient has been found when a moderating agent is added to the gas. This effect has been explained in terms of the velocity dependence of annihilation rate.

For methane and deuterated methane the annihilation rate λ again shows a linear dependence on the pressure. However, the results varied widely from gas cylinder to gas cylinder, suggesting an impurity effect. The observed values of annihilation rate per unit pressure for the two gases consequently overlap and it was not possible to determine the difference between the two.

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Finally I wish to record my gratitude to N.R.C. for a summer grant and for financing the project and to the University of Toronto for providing me with the job of Student Demonstrator.

CHAPTER.I.

Introduction

The positron was discovered by Anderson (A-32) in a cloud chamber photograph from cosmic ray shower. The discovery was also made independently by Blackett and Occhianlini in 1933 (B-33) who indetified the particle with the "positive hole" in the continuum of negative energy states postulated by Dirac (D-30). Solving the relativistic equation for the energy of the electron, Dirac obtained both positive and negative solutions

$$E = \pm \sqrt{p^2 + mc^2}$$

The negative energy solution corresponds to a positive energy solution in which the electron has a charge +e instead of -e. The connection between positive and negative energy state is described by Dirac in his "Hole theory" in which it is assumed that all negative energy states are normally occupied by electrons which are unobservable. When an electron makes a transition from a negative energy state to a positive energy state a hole is left which is observable as a positron, and the electron in the positive energy state may make radiative transition into a "hole" resulting in the disappearance or annihilation of the positron and electron and the appearance of electromagnetic radiation of total energy

$$E = 2m_oc^2 + E_+ + E-$$

where m_o is the rest mass of the electron and E_+ and E_- are kinetic energies associated with the positron and electron respectively.

The conservation of momentum requires atleast two quanta to be emitted unless an interaction with another electron or nucleus occurs. Symmetry consideration (Y-50) requires an even number to be emitted in case of opposite spins of annihilating pair, and an odd number (atleast three) in case of parallel spins.

Production of positrons

Positron electron pairs can be created by electromagnetic radiations in the vicinity of an atomic nucleus. To do this the energy of electromagnetic radiations must be sufficient to cause a transition from the negative energy state $-m_oc^2$ to a positive energy state $+m_oc^2$. Thus a minimum energy of $2m_oc^2$ is required to produce an electron positron pair. The presence of a nucleus is required for energy and momentum conservation in the process of creation. The positrons are thus created in cosmic radiation and whenever high energy gamma rays pass through matter.

Positrons are also emitted by certain radioactive nuclides which have neutron deficiency. For most experiments on positron annihilation such sources are very useful.

Fate of positrons in gases

In many experiments Na-22 is a most convenient source. It has a half life of 2.58 years, and the positrons have an energy distribution with a maximum of 0.544 Mev. (E-54). These positrons are passed through a substance where they lose almost all their energy by ionising collisions with atoms and molecules of the medium. Tao, Green and Celitans (T-63) have estimated the thermalization time for positrons in argon gas by separating the energy of positrons into three regions:

(i) $E > 0.01$ Mev.

(ii) $0.01 \text{ Mev} > E > E_{exc.}$

(iii) $E < E_{exc.}$

where $E_{exc.}$ is the first excited level of argon atom. while this thesis is not primarily concerned with argon, it will be convenient to consider argon in the following description.

In stage (i) the positron annihilation probability is less than few percent. The slowing down time is estimated to be about $0.025 \tau_f$ where τ_f is the Dirac free annihilation time, amounting, for example to 55.2 nsec. for argon gas at 5 atmosphere pressure.

In stage (ii) the slowing down time is estimated to be $0.32 \tau_f$. Annihilation is less than 30% probable.

In stage (iii), 11.6 ev to 9.0 ev.* the probability of positron capturing an electron from an argon atom and forming a bound state is almost 100%. The bound state is analogous to a hydrogen atom with positron in place of proton. This concept was first given by Mohorovicic (M-34) and bound positron electron pair was given the name Positronium by Ruark (R-45).

A positron which reaches energy less than 9.0 ev. in argon (and many do so by escaping the energy range 11.6 ev - 9.0 ev altogether) can no longer form positronium and it will continue to slow down losing energy in elastic collisions until it annihilates.

Similar conclusions can be reached for other monatomic gases. In polyatomic gases the situation at low energies (region iii) is somewhat different because inelastic scattering collisions occur with high probability. Hence the thermalization time is short and most of the positrons would annihilate after reaching thermal or nearly thermal energies.

Paul and St.Pierre (P-63) have reported that in these gases the annihilation rates are from 3-700 times faster than those obtained by Dirac formula:

$$\lambda = n\sigma v = n\pi r_0^2 c \ \text{Sec}^{-1}$$

---

* The significance of the level 9.0 ev is that it equals the ionisation potential of argon less that of positronium.

where n is the number of electrons per unit volume and r_0 the classical electron radius.

To explain the enormous annihilation rates in the heavier gases these authors have suggested the possibility of the formation of positron-molecular ions, which would annihilate in about 10^{-9}sec. Therefore the observed annihilation rate would represent a combination of the rate of capture of positrons to form bound states and the direct annihilation rate. Since the total observed rate is much greater than the direct annihilation rate at low pressures, one is essentially observing a phenomenon which depends on the process

$$X + e^{+} \longrightarrow Xe^{+} + Q$$

where X is the capturing molecule, and Q is the binding energy of the compound Xe^{+}. The energy Q may be taken up in a rotational or vibration-rotational transition of the molecule, or could be radiated, or dissipated collisionally. The latter process would require a virtual state to exist for about 10^{-10}sec. in a gas at one atmosphere, and since all the experiments described in this thesis were for overall pressures above one atmosphere, collisional de-excitation would take preference over optical emission, which is generally slower than 10^{-8}sec. for low energy transitions. Returning now to the vibration-rotation transitions, the number of available channels for such transitions should be greater for molecules with high densities of rotation-vibration states. The rapid increase of annihilation rate with molecular weight suggested that vibration-rotation transitions were playing a major role. It was suggested by Dr.H.L.Welsh that a comparison of annihilation rates in methane and deuterated methane (CD_4) would reveal any such tendency, since the two molecules are essentially identical in their electronic structure and electric polarizability, but have markedly different moments of inertia. It was therefore decided to compare the "free" positron annihilation rates in these two substances. Failure to observe any difference would not necessarily mean that positrons were not being captured, but rather that if they are captured they do not simultaneously cause a vibration-rotation transition.

We must also bear in mind that it has not yet been shown that the capture of positrons occurs at all by the molecules in question, even though bound states exist. Recent calculations of Massey (Private communication) indicate that if electric polirazition of argon is taken into account in the theory of positron collisions with the free atoms, the annihilation rate for very low velocity positrons is markedly higher than if the polarization is neglected (the reverse is true for positrons of about 3-9 ev in argon). Thus it is still possible that a very high direct annihilation rate could be demonstrated theoretically for hydrocarbons. However, the recent communication of Gold'anskii and Sayasov (G-64) favours the formation of bound states. (see Appendix A).

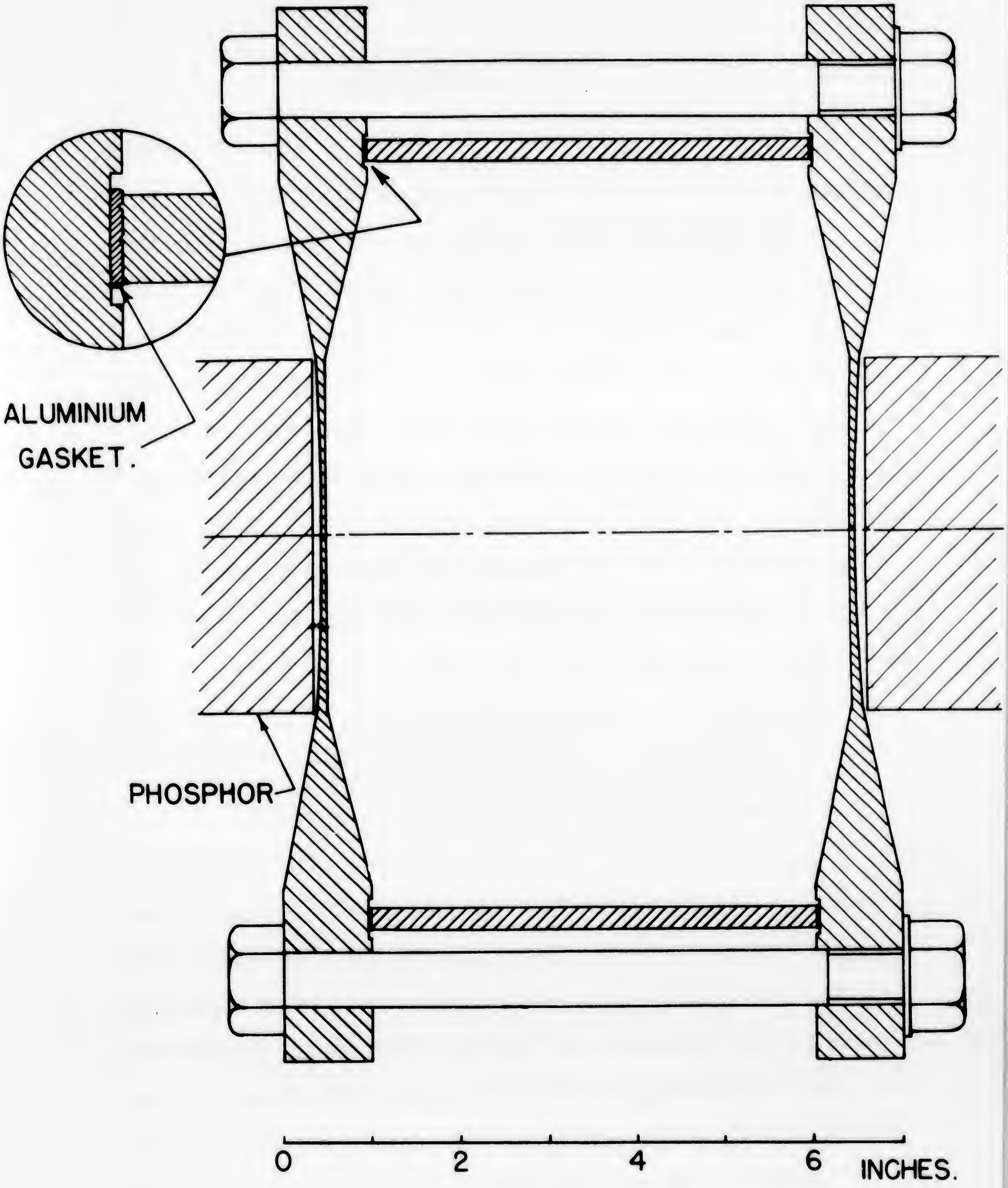


FIG. 1 STAINLESS STEEL PRESSURE CHAMBER.

CHAPTER.II.

Method

To measure the "free" lifetime of the positrons in the mixture of gases, Na-22 was used as a source of positrons. This gives positrons having energy distribution characteristic of β-decay. Na-22 decays into an excited state of Ne-22 which goes to the ground state in less than 10^{-11} ses. giving 1.28 Mev γ-rays. Thus emission of every positron is marked by the emission of a γ-ray of 1.28 Mev. The 1.28 Mev γ-ray may therefore,be regarded as signal for the creation of a positron. The positron subsequently annihilates into two γ-rays of 0.51 Mev. The time interval between the 1.28 Mev γ-ray and the annihilation γ-ray gives the lifetime of individual positron.

The source was mounted in the centre of a stainless steel cylinder 8 inches in diameter and 6 inches long (figurel) in which the gas under study could be compressed upto 10 atmospheres. The gas chamber was evacuated every time before the gas under study was introduced. The vacuum system was designed and constructed during the course of these experiments. The pressure of the gases were measured using (Wallace and Tiernan Model No.FA 233) pressure gauges of three differnt ranges (0-400, 400-800, 0-7600 mms Hg).

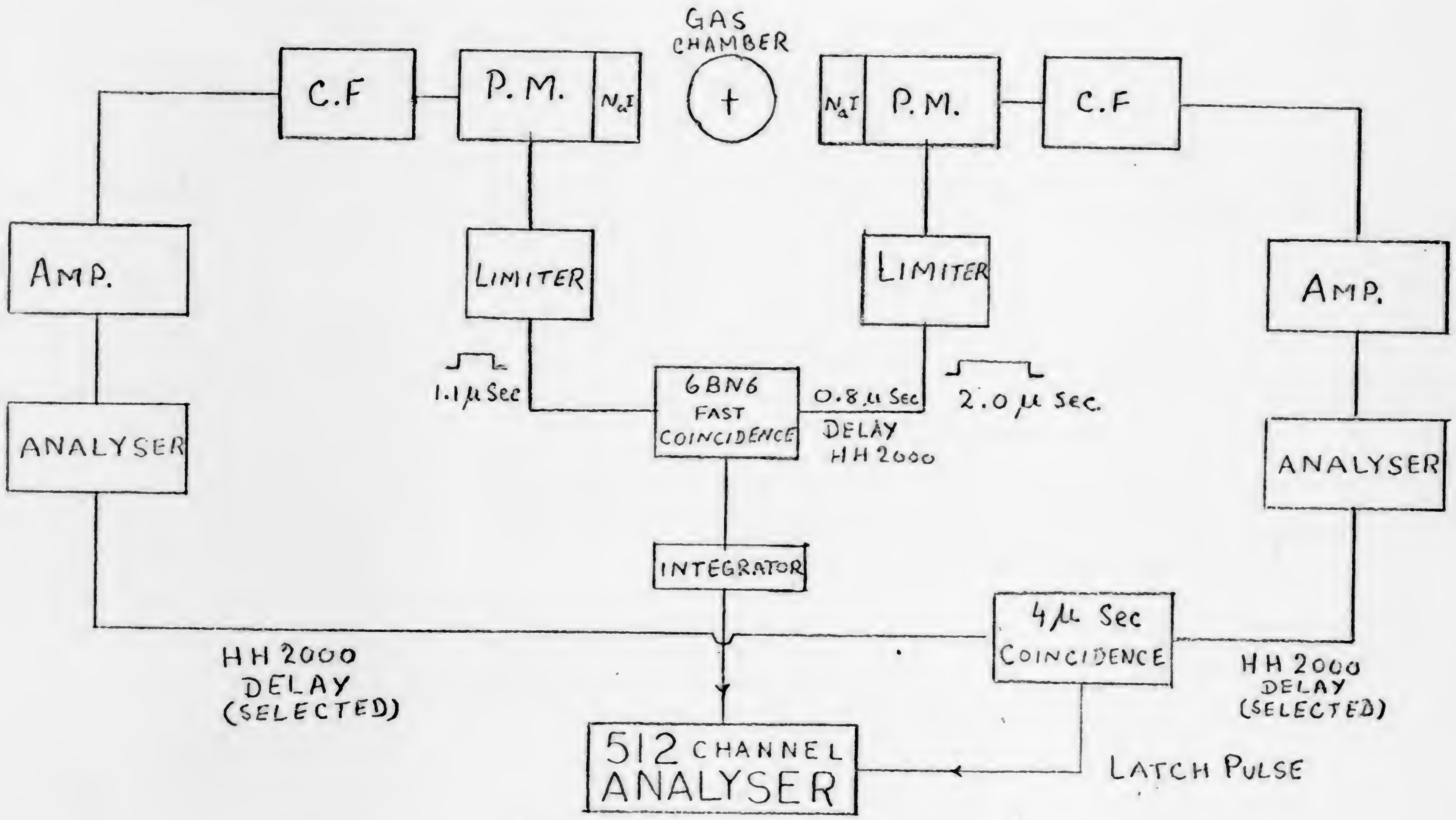


FIG.2. BLOCK DIAGRAM OF ELECTRONIC CIRCUIT

Two NaI(Tl) detectors mounted on large diameter photomultipliers (Phillips XP-1040 and RCA 7046) were placed face to face, with the gas chamber lying between them. By the pulse amplitude selection the two detectors were set to detect the 1.28 Mev γ-ray and annihilation γ-ray respectively. Two output signals were extracted from each of the two photomultipliers. A time pulse from each photomultiplier,is fed to a coincidence circuit (timesorter) which counts coincidences over a range of different delays. These time delays are converted to pulse amplititude by a time to pulse height converter. For pulse sorting a Nuclear Data model "ND 120" 512 channel analyser was used. Another pair of pulses obtained from the photomultipliers (side channel pulses) was used for gamma ray identification and also to produce a gate pulse which controls the time sorting device. The electronic circuits used in these experiments were the same as were used by Paul and St.Pierre (P-63). The block diagram of the entire circuit is given in figure 2.

To calibrate the timesorter six cables of RG11/U, each 100 feet long, were cut. The delays of these cables were measured by using a Hewlett Packard "Time Domain Reflectometer model 1415A". These measurements were checked carefully by a standing wave method employing

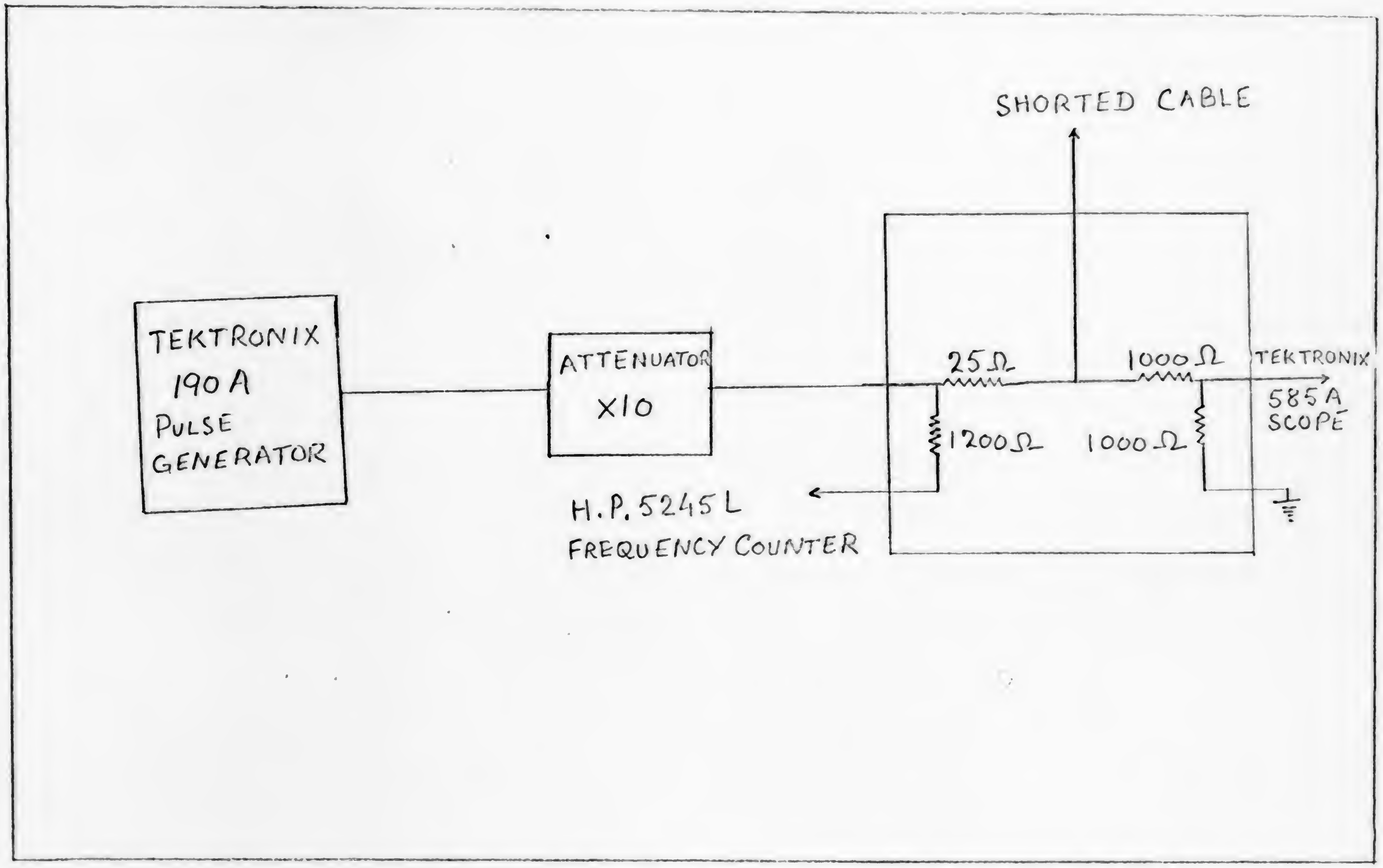
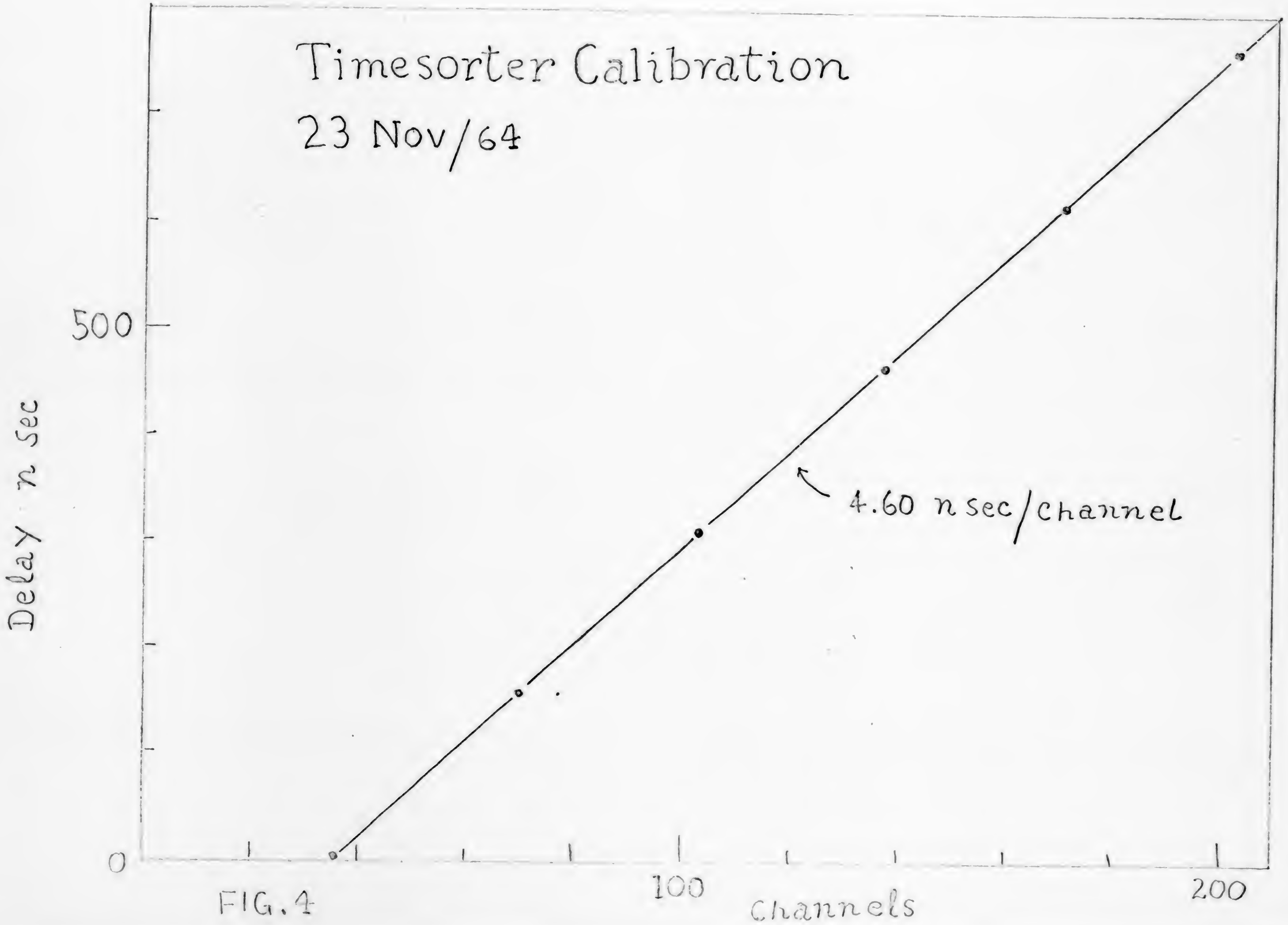


FIG. 3. Circuit For Calibration of RG/11U Cable.



Timesorter Calibration

23 Nov/64

FIG. 4

a variable frequency generator and frequency counter (figure.3). The values of the delays measured by these two methods agreed to better than ± 0.05 % apart from a systematic difference 0.4 %. Such a difference is only to be expected beacuse of the Time Domain Reflectometer, for which the manufacturer claims only an absolute accuracy of ± 5%. We have thus demonstrated that the particular Reflectometer used is accurate about ± 0.5% or better.

Pulses from a home made mercury pulser having a fast rise time were than used to calibrate the timesorter by the method described in references (P*58) and also (G-62). The calibration also served to check the linearity of the timesorter (figure.4.). In our most recent work the linearity of the timesorter over the range 0-465 nsec has been better than that of the ND-120 analyser itself. We therefore felt justified in ignoring any slight non-linearity which may be present.

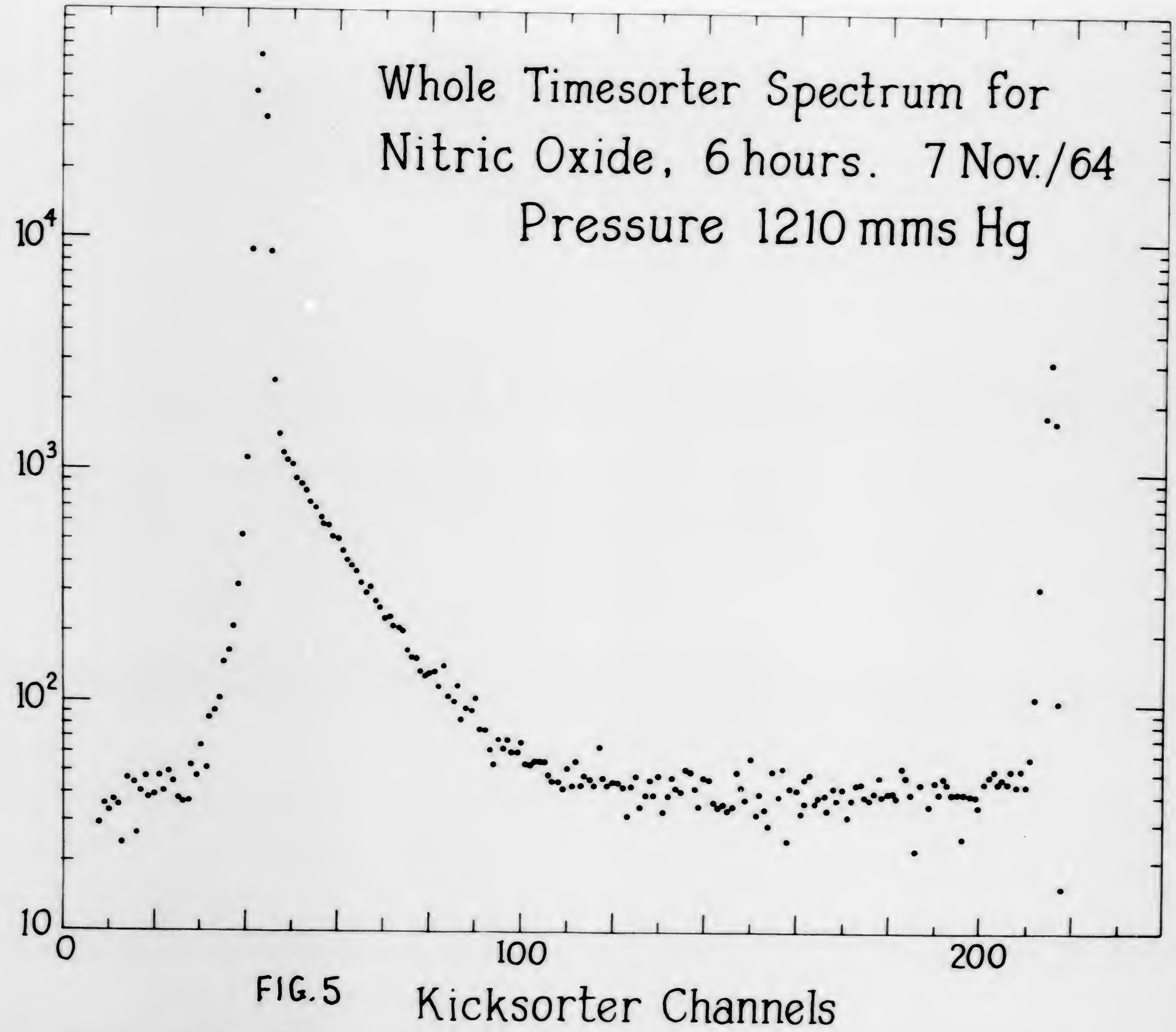

Whole Timesorter Spectrum for
Nitric Oxide, 6 hours. 7 Nov./64
Pressure 1210 mms Hg
10^4
10^3
10^2
10
0
100
200
Kicksorter Channels

FIG. 5

CHAPTER.III.

Experimental Results

The annihilation rates of free positrons were measured as a function of pressure in the following gases and mixtures of gases:

1. Nitric Oxide
2. Nitric Oxide + Argon
3. Methane + Nitric Oxide + Argon
4. Deutrated Methane + Nitric Oxide + Argon
5. Methane + Nitric Oxide + Argon

(the pressures of the underlined gases were varied)
A typical time spectrum is shown in figure 5. All the data curves were processed by a three parameter least square fitting procedure, described by Paul and St.Pierre (P-63),using the Toronto University IBM 7090 Computer.

In sets 2-5 of the measurements Nitric Oxide was merely used to quench Ortho-Positronium,so that this normally long lived component would not appear in the spectrum of delayed annihilations. (The quenching of Ortho-Positronium by Nitric Oxide is a well known phenomenon see H-61.). In sets 4 and 5 the function of the Argon gas is merely to slow down the positrons within the distance of the order of the dimensions of the vessel, thus reducing the number of annihilations in the walls.

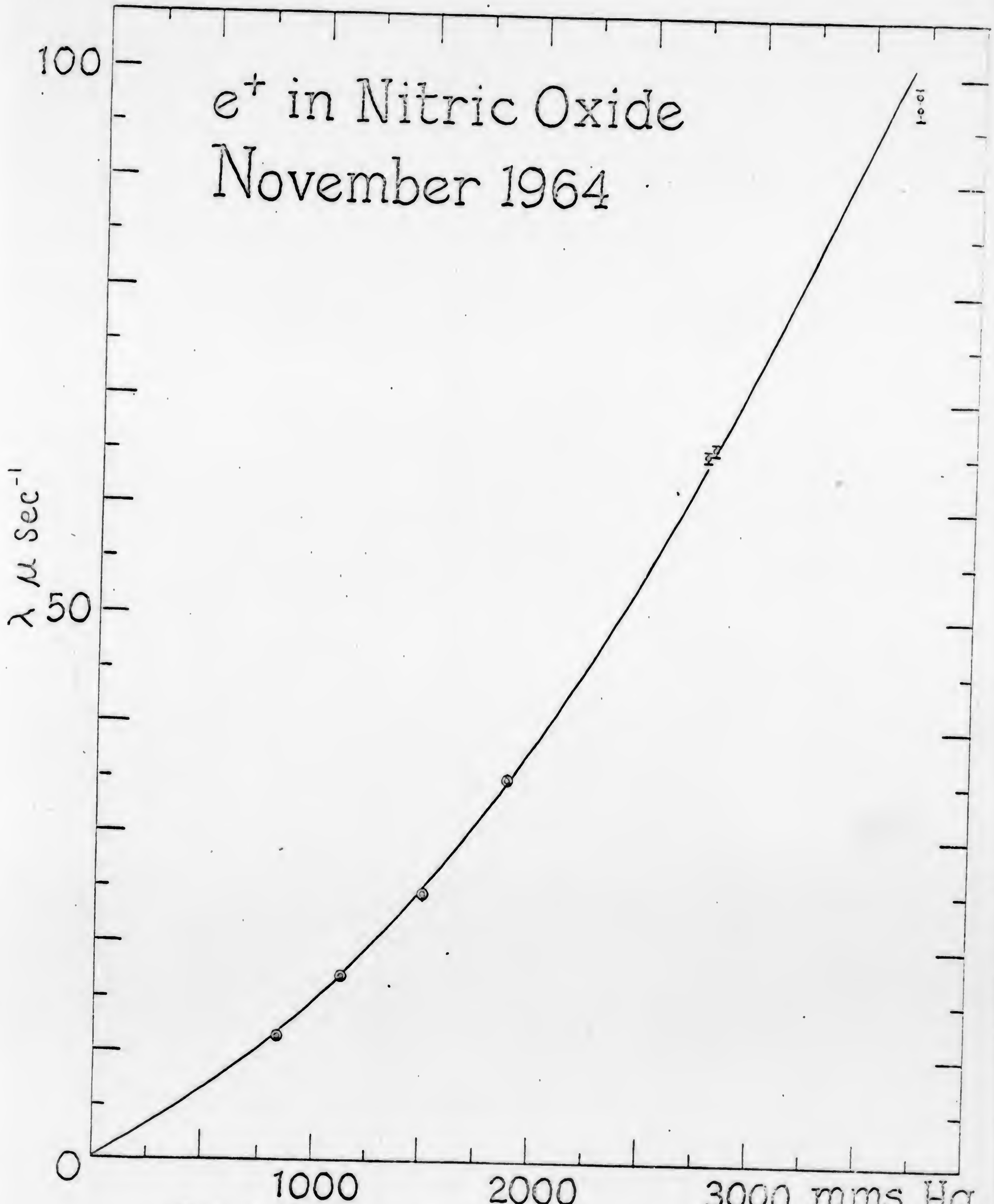

e$^+$ in Nitric Oxide
November 1964
λ μ Sec^{-1}
100
50
0
1000
2000
3000 mms Hg
Pressure

FIG. 6.

Set 1. Lifetime measurements in Nitric Oxide

Free positron lifetimes were measured at eight different pressures of nitric oxide in the pressure range of 843 mms Hg - 3680 mms Hg. The time spectrum for each run was taken for 400 minutes. The results of these measurements are tabulated in table 1. and are given graphically in figure 6.

Table 1.

Runs	Nitric Oxide mms Hg at standard temp.	sec^{-1}	Statistical error in sec^{-1},
1	3680 ± 5	98.2	1.0
2	2763	65.0	0.4
3	1869	35.0	0.2
4	1125	17.1	0.1
5	2799	65.7	0.4
6	4496	94.5	5.2
7	1495	24.7	0.1
8	844	11.4	0.2

Set 2. Nitric Oxide + Argon

In this set of runs a fixed amount of about 282 mms Hg of nitric oxide was used and the pressure of

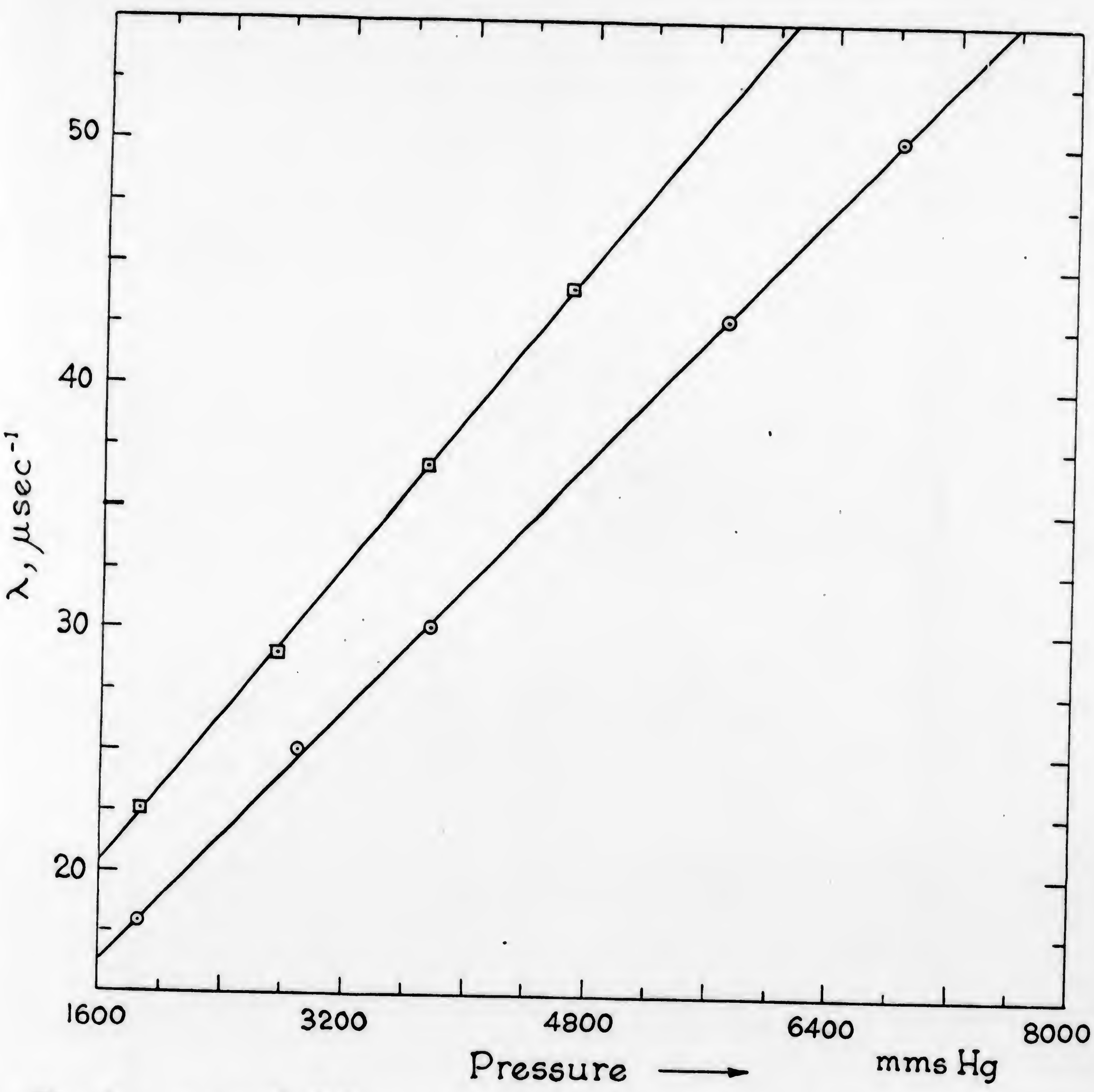


Fig. 7 ⊙ Annihilation rates in argon with ~ 280 mms Hg Nitricoxide
⊡ Annihilation rates in argon with ~ 140 mms Hg methane + ~ 280 mms Hg Nitricoxide

argon was varied from 1860 mms Hg - 6817 mms Hg in five steps. The results obtained are tabulated in table 2. and illustrated in figure 7.

Table 2

Runs	Nitric Oxide mms Hg at standard temp.	Argon mms Hg at stan. temp.	$\lambda, \mu sec^{-1}$	Statis. error $\lambda, \mu sec^{-1}$
1	279 ± 2	1860 ±5	17.9	0.2
2	282	3760	30.1	0.3
3	283	2892	25.0	0.3
4	284	5688	42.7	0.3
5	279	6817	50.1	0.2

Set 3. Methane + Nitric Oxide + Argon

In this set of runs fixed amounts of methane and nitric oxide, ~140 mms Hg and ~280 mms Hg respectively were used. The pressure of argon was varied from 1868 mms Hg - 4664 mms Hg (table 3). These measurements were made for comparison with those of set 2. The results are given in figure 7.

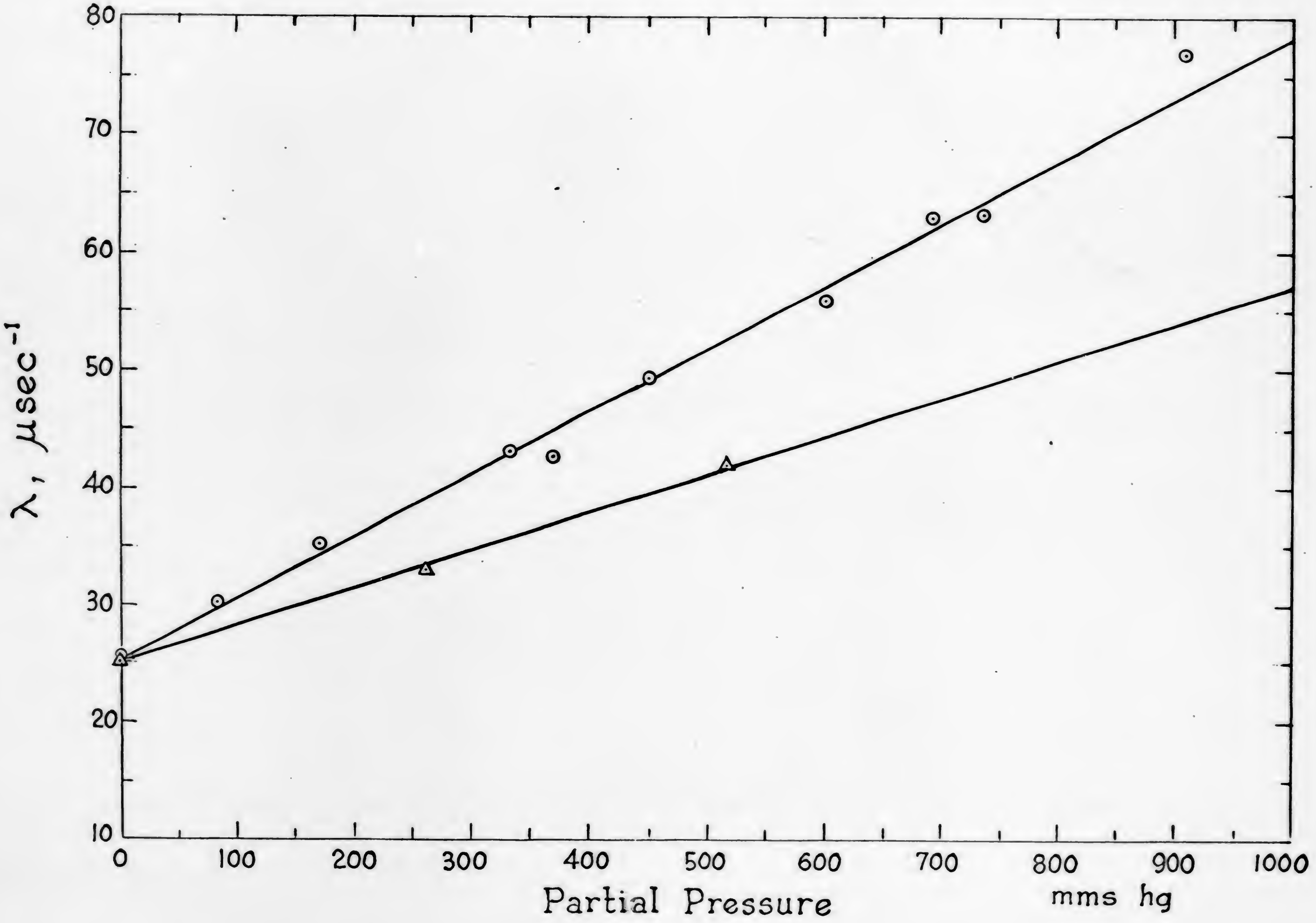


Fig. 8 Annihilation rates in dentrated methane △ 1st group ⊙ 2nd group

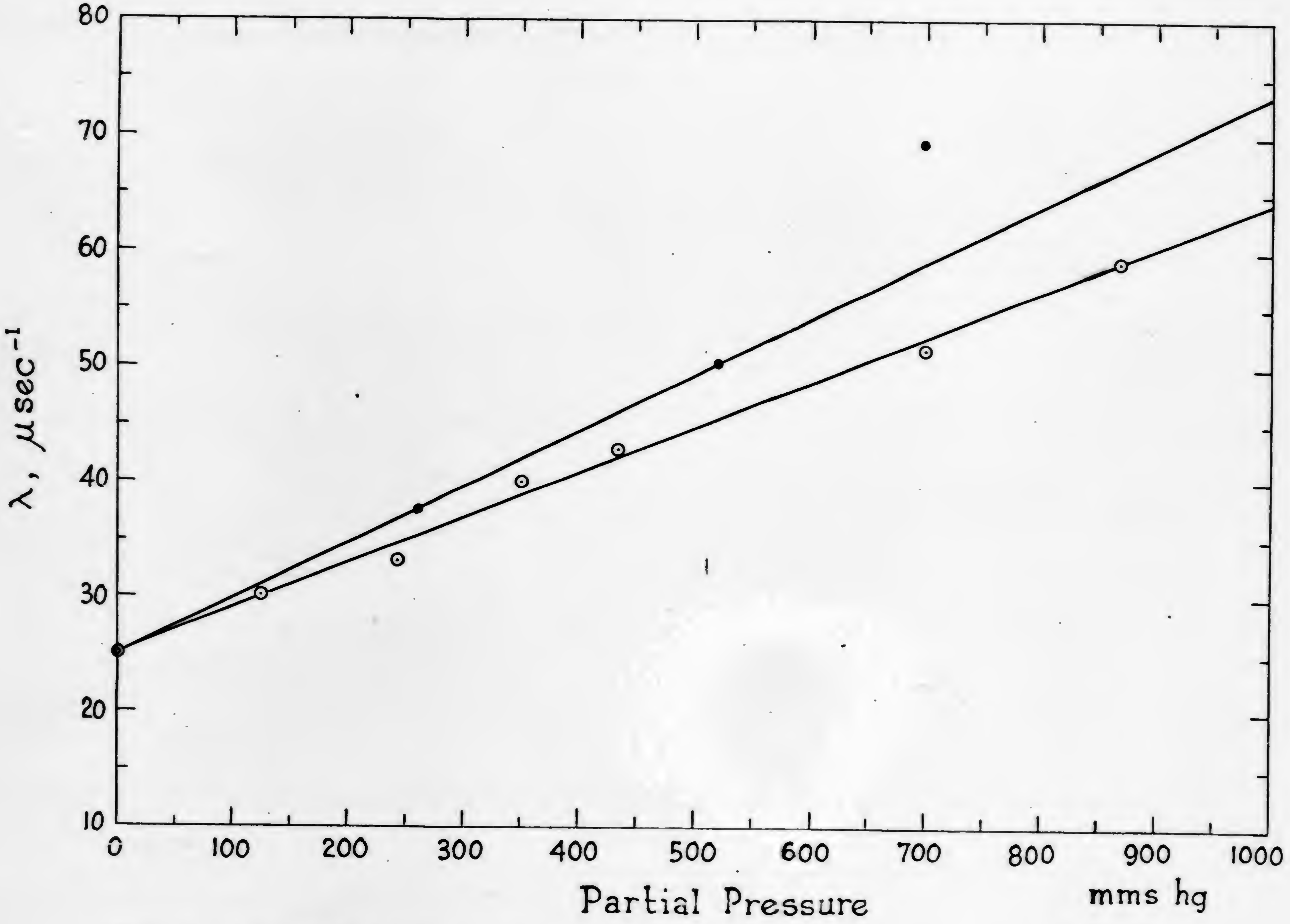


Fig. 9 Annihilation rates in methane • 1st group ⊙ 2nd group

Table 3.

Runs	Methane mms Hg stan. temp.	Nitric Oxide mms Hg stan. temp.	Argon mms Hg stan. temp.	$\lambda, \mu sec^{-1}$	Statis. error $\lambda, \mu sec^{-1}$
1	140 ± 2	280 ± 2	1868 ± 5	22.6	0.1
2	139	279	2790	29.2	0.2
3	140	280	3728	36.7	0.2
4	141	281	4664	44.2	0.3

Set 4. Deuterated Methane + Nitric Oxide + Argon &

Set 5. Methane + Nitric Oxide + Argon

In these sets fixed amounts of argon and nitric oxide ~2840 mms Hg and ~280 mms Hg respectively were used. The pressures of the hydrocarbons were then varied in order to study the difference of annihilation rates in these two gases, in similar circumstances. The results of these measurements are given in tables 4 & 5 and are shown graphically in figures 8 & 9.

Table 4.

First Group

Runs	Deuterated Methane mms.Hg. S.T.	Nitric Oxide mms.Hg. S.T.	Argon mms.Hg. S.T.	λ, μ sec^{-1}	Statistical error λ, μ sec^{-1}
1.	261 ± 2	279 ± 2	2827 ± 5	33.4	0.1
2.	561	276	2799	42.2	0.2
3.	0	282	2891	25.0	0.4
	Second Group				
1.	692 ± 2	280 ± 2	2833 ± 5	62.9	0.3
2.	600	277	2809	55.9	0.4
3.	332	280	2833	42.9	0.2
4.	170	278	2812	35.1	0.2
5.	86	280	2833	29.9	0.2
6.	735	277	2809	62.8	0.5
7.	370	280	2833	41.8	0.2
8.	902	279	2827	76.8	0.5
9.	450	278	2821	49.1	0.2
10.	0	282	2891	25.0	0.2

Table 5.

First Group

Runs	Methane mms. Hg. S.T.	Nitric Oxide mms. Hg. S.T.	Argon mms. Hg.	$\lambda, \mu\, sec^{-1}$	Statistical error $\lambda, \mu\, sec^{-1}$
1.	262 ± 2	279 ± 2	2827 ± 5	37.8	0.2
2.	518	277	2803	50.1	0.4
3.	697	281	2842	69.5	0.9
4.	0	283	2891	25.0	0.2
	Second Group				
1.	866 ± 2	280 ± 2	2845 ± 5	59.0	0.4
2.	433	280	2845	42.9	0.3
3.	699	281	2851	51.5	0.2
4.	349	281	2848	40.0	0.3
5.	244	282	2856	33.1	0.2
6.	122	280	2845	30.2	0.2
7.	0	282	2891	25.0	0.2

The maximum impurity in CD_4 specified by the Manufacturers is 0.5% air. There are no other hydrocarbons present, while the isotopic impurity is 1% of hydrogen (atomic).

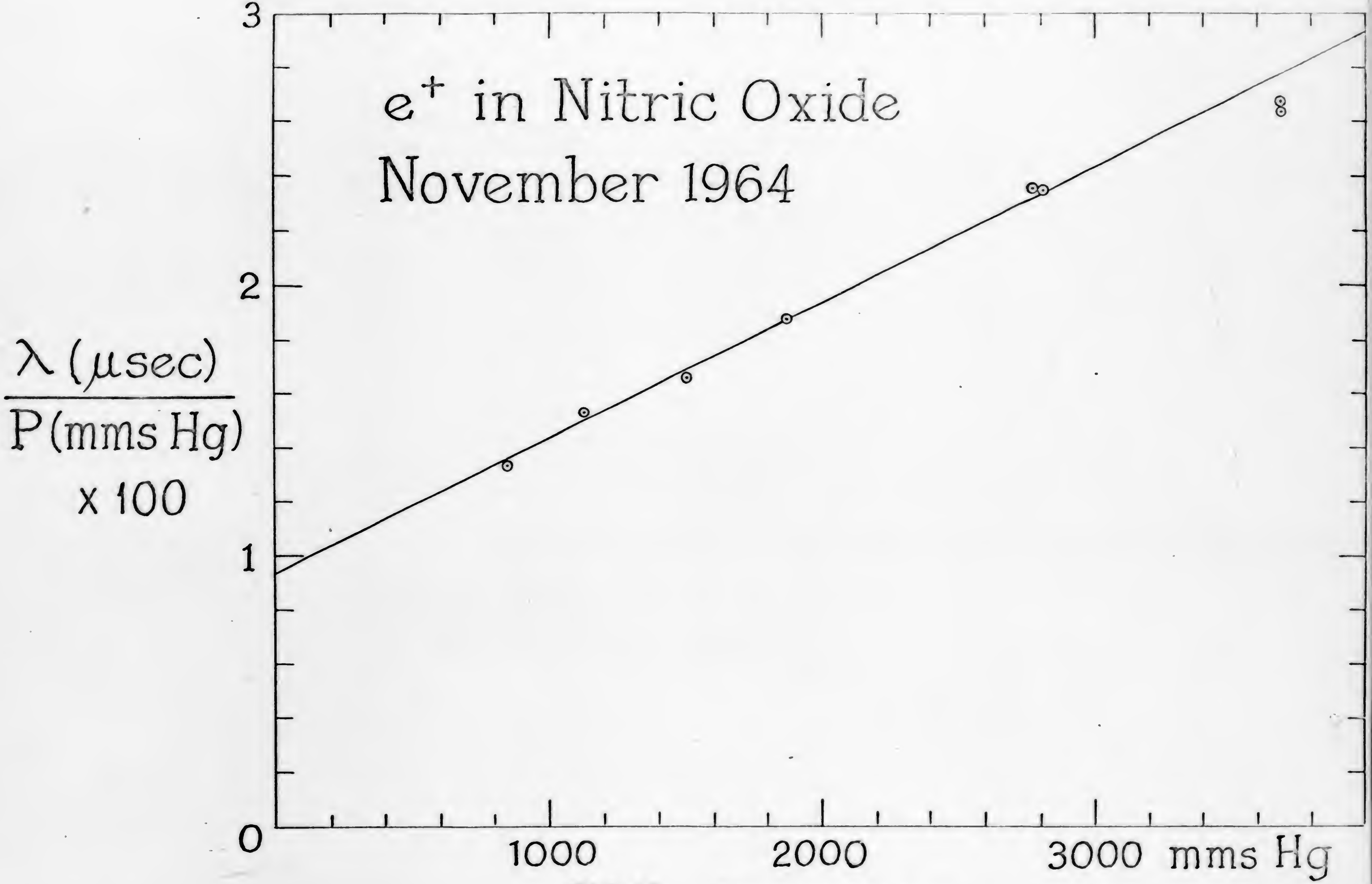

e^+ in Nitric Oxide
November 1964
$\frac{\lambda\,(\mu sec)}{P\,(mms\ Hg)}$ x 100
3
2
1
0
1000
2000
3000 mms Hg
Pressure

FIG.10.

CHAPTER.IV.

Discussion of the results

1. Measurements in Nitric Oxide:

Paul and St.Pierre (1963) used nitric oxide as one component of their gas mixtures and obtained an approximate value (unpublished) for the annihilation rate in this gas. They experienced some difficulty in determining a constant value λ/p, which led Paul (1963 unpublished) to make some measurements on nitric oxide alone.

The plot of annihilation rate against pressure obtained shows clearly that the annihilation rate λ has a quadratic dependence on the pressure of the form

$$\lambda = ap + bp^2.$$

This is the first instance of nonlinear dependence of λ on pressure which has been found. These measurements have been carefully repeated (figure 6). To determine the values of "a" and "b" a plot is made for λ/p against p (figure 10). From this plot is obtained

$$a = (9.3 \pm 0.5) \times 10^{-3}\ \mu\text{sec}^{-1}/\text{mms.Hg}$$

$$b = (5.0 \pm 0.3) \times 10^{-6}\ \mu\text{sec}^{-1}/(\text{mms.Hg})^2$$

It may be emphasized, however, that the value of "b" appears to be highly sensitive to the experimental conditions, and may indicate a strong dependence of "b" on impurities. Nevertheless two sets of data obtained prior to the results given in the previous chapter have given consistent values of "a".

The quadratic dependence on pressure can be explained on one of the several theories.

(i) Capture effect into a bound state.

$$NO + e^+ \longrightarrow (NO^+)^*$$

followed by

$$(NO^+)^* \longrightarrow NO + e^+$$

or $$(NO^+)^* + NO \rightarrow \underset{\substack{\downarrow \\ \text{rapid annihilation}}}{NO^+} + NO + Q$$

Q is the binding energy of the positron in NO^+. The positron in this NO^+ bound state is expected to annihilate fairly rapidly,with a mean life of the order of 10^{-9}sec.

(ii) Dimerization effect $(NO)_2$.

According to this theory we assume that the annihilation rate λ is equal to the sum of annihilation rate in NO, plus the annihilation rate due to the dimer $(NO)_2$.

i.e. $$\lambda = \lambda_{(NO)} + \lambda_{(NO)_2}$$

and $$\lambda_{NO} \propto p$$

$$\lambda_{(NO)_2} \propto p^2$$

The p^2 dependence of $\lambda_{(NO)_2}$ is expected beacuse the concentration of $(NO)_2$ will vary as p^2. (This follows from the kinetics of the formation and breakup of $(NO)_2$

If we have a mixture of NO and $(NO)_2$. Then in the equilibrium the concentration of the dimers is proportional to the square on the concentration of monomers,

i.e. $C_{(NO)_2} = KC^2_{NO}$

or $C_{(NO)_2} = (\text{pressure})^2$ approximately.

This approximation is accurate at low $C_{(NO)_2}$ values, such as obtained under our experimental conditions. The constant K depends on temperature in a manner involving the factor $e^{Q/KT}$ where K is the Boltzmann's constant and Q is the dissociation or activation energy for the breakup of the dimer into NO + NO. The value of Q = 3710 Cal/mol or 0.1624 ev per molecule, has been determined experimentally by Smith and Johnston (S-52).

On this theory the parabolic contribution to the curve of λ versus p is merely due to a large cross-section for annihilation of slow positrons with very few dimers which are present. The mechanism for such a large annihilation cross-section could be similar to that for medium weight hydrocarbons (see for example Appendix A). We do not attemt here to explain such large annihilation rate, but merely compute the order of magnitude of the annihilation cross-section, which would be necessary to account for our observed parabolic pressure dependence on a dimerization picture.

$$\lambda_{(NO)_2} = n\sigma_{(NO)_2}v$$

where n is the number of $(NO)_2$ molecules per cm^3, and

$$\sigma_{(NO)_2} = n_{eff}\sigma$$

where $n_{eff.}$ is the number of electrons per $(NO)_2$ molecules which are effective in annihilating the positron, and σ is the Dirac cross-section

$$\text{i.e}\quad \sigma = \pi r_0^2 c/v$$

However the number of dimers present is given by

$$n = \nu_p p^2$$

$$\therefore\quad \lambda_{(NO)_2} = \nu_p p^2 n_{eff}\sigma v = bp^2$$

$$= \nu_p p^2 n_{eff} \times .748 \times 10^{-14}\ Sec^{-1}$$

$$\therefore\quad b = \nu_p n_{eff} \times .748 \times 10^{-20}\ \mu\,Sec^{-1}/(mms.\,Hg)^2$$

$$\text{-----}\ (i)$$

Supposing now that the fraction of dimers present at one atmosphere is 10^{-5}, then number of $(NO)_2$ molecules per $cm^3 = 10^{-5} \times L$, where L is the Loschmidts number.

$$\therefore\ \nu_p = 10^{-5}L/(760)^2$$

Substituting in equation 9i) and using experimental value of b

$$n_{eff.} \approx 1.43 \times 10^6 \quad \text{electrons/molecule.}$$

The highest value of $n_{eff.}$ by Paul and St.Pierre (P-63) is about 22000, for carbon tetrachloride. Therefore $n_{eff.}$ would appear to be anomalously high on this picture unless we have drastically underestimated the fraction

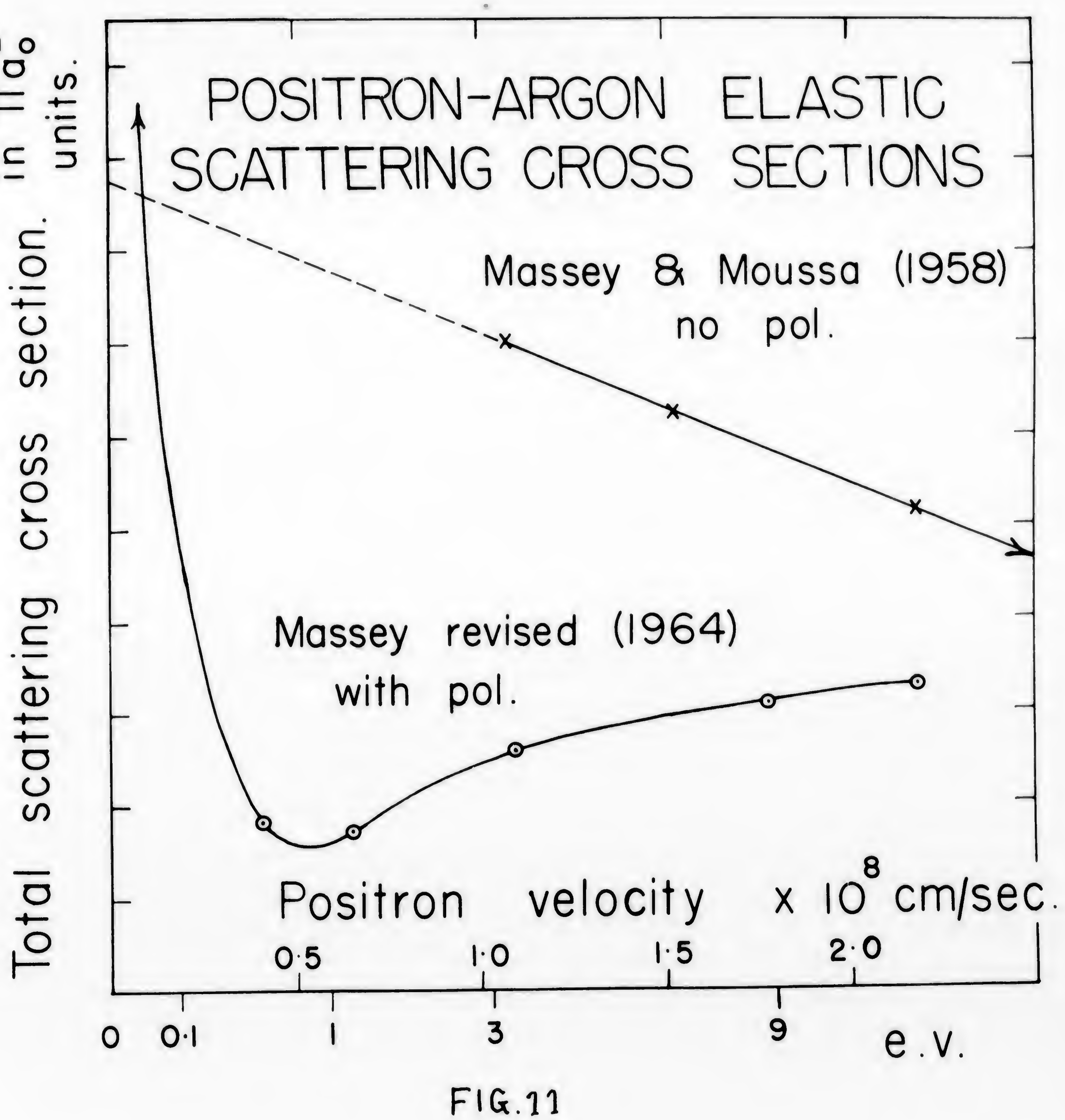

POSITRON-ARGON ELASTIC SCATTERING CROSS SECTIONS
Massey & Moussa (1958) no pol.
Massey revised (1964) with pol.
Total scattering cross section. in πa_0^2 units.
Positron velocity x 10^8 cm/sec.
0·5
1·0
1·5
2·0
0
0·1
1
3
9
e.v.

FIG.11

of dimers which is present.

2. Measurements in Nitric Oxide + Argon and Methane + Nitric Oxide + Argon

The plots of annihilation rate λ against partial pressures of argon (figure.7) are straight lines, as has always been found with gases except for nitric oxide (cf. secl). The curves of figure 7 are similar except that the upper curve has a slightly steeper slope. (The difference in intercepts is merely due to the positron collisions with methane). The greater slope of the upper curve can be understood as a thermalization effect. It is to be expected that the average velocity of slow positrons in the gas will be greater than thermal beacuse the slower positrons are annihilated preferentially. Methane moderates the positrons more rapidly,so that the average positron velocity is reduced towards thermal energies. A consequence of the lower positron velocities is an increased λ .There is already some published experimental evidence that the annihilation rate of positrons in argon rises as the positrons slow down below about 1 ev (F-64, T-64, P-64). The latest calculations Massey (1964 private communication) also bears this out (figure 11). " It seems that a Ramsauer effect occurs for positrons, and annihilation rate shows this effect in a smeared out fashion".

3. Measurements in Deuterated Methane + Nitric Oxide + Argon and Methane + Nitric Oxide + Argon

The annihilation rates λ obtained with deuterated methane are plotted against their partial pressures (figures 8 & 9). The plots are straight lines, but the sol slopes in each figure differ according to the gas samples. (Two cylinders of each type of gas were purchased at different times).

We therefore suggest that annihilation rates in these gases are very sensitive to impurities present. Thus it is difficult to measure the difference of annihilation rates in CD_4 and CH_4. However, the greatest observed difference of annihilation rates in these two gases is about 14%. The annihilation rate in CD_4 being higher, while the maximum difference of annihilation rate between two samples of the "same" gas is 10%. It is therefore not possible to determine whether the annihilation rates of positrons in CD_4 and CH_4 are different.

Gol'danskii and Sayasov (G-64) postulate a resonance capture phenomenon to explain the decay of positrons in CH_4, and suggest an annihilation rate from a bound state having a life-time of about 10^{-8} sec. From an experimental point of view the Gol'danskii and Sayasov's theory is incomplete. A phenomenological theory of the rates of processes appropriate to Gol'danskii and Sayasov has therefore been been developed by Paul (Appendix B). From it we are able to deduce the pressure dependence of annihilation rate in CH_4 as a function of five parameters. The values of several of these parameters can only be guessed,but using the theory we are able to make rough estimates of λ_f ,the bound state formation rate from experimental results. It is this parameter which is expected to depend on the rotation level spacing of the molecule,and which should give rise to an enhanced annihilation rate in CD_4 as compared to CH_4. Taking a partial $n_{eff.}$= 20 for direct annihilation in CH_4 the value of λ_f is about 92% of the total observed annihilation rate. Hence we may expect the annihilation rate in CD_4 to be higher than in CH_4. Despite experimental difficulties (impurity effects) it is improbable that our observed results could be fitted to the ideas just presented unless λ_f is much smaller than has been supposed and the direct annihilation rate much larger. At present we are therefore inclined to fit to our results smaller values of C_f (Appendix B) and a larger value of direct annihilation rate. This would imply that the theory of Gol'danskii and Sayasov applies only to a much smaller fraction of the positrons than 92%.

The results discussed in this section have not been corrected for the second order effects discussed in section 2 which are due to the presence of argon.

Appendix A

In this appendix Gol'danskii and Sayasov's paper on "Resonance annihilation of of positrons upon collision with molecules and atoms" is presented, since this paper is not available in English.

The lifetime of positrons in a number of hydrocarbons has been shown to be several orders shorter than those obtained by Dirac formula: $r_p = 1/\sigma_p vn$, $\sigma_p = r_0^2 Z_b\, c/v$ --(1) where r_0 is the classical electron radius, n the number of molecules/cm^3, Z_b the number of valence-shell electrons and v the velocity of positrons. An interpretation of this phenomenon is given below.

Positron is slowed down to the energies of the order of 0.1 ev in the time considerably shorter than the Dirac annihilation time. If a losely bound positron-molecule state having binding energy $\varepsilon \sim 0.1$ ev exists, then the positron is annihilated subsequently on the molecule, as in the complete resonance manner, with the probability considerably exceeding Dirac probability.

In other words, an intermediate long-lived positron-molecule state develops in the process of scattering with one of the disintegration channels of this state being annihilation. The cross-section is given by Wigner type formula:

$$\sigma_a = \frac{2\pi}{K^2}\sqrt{\frac{E}{\varepsilon}}\,\frac{\Gamma_a}{E+\varepsilon}\,, \quad K^2 = \frac{2m_e E}{\hbar^2} \text{ --- (2)}$$

is the level width in relation to annihilation. The ratio of the cross-sections of resonance and Dirac annihilation, equal to the obeserved ratio of annihilation rates, may be represented by the formula:

$$\frac{\sigma_a}{\sigma_p} = 2\left(\frac{\hbar c}{e^2}\right)^2 \frac{1}{Z_6 \beta_0} \frac{\Gamma_a}{E+\varepsilon} = 3.76 \frac{10^4}{Z_6 \beta_0} \frac{\Gamma_a}{E+\varepsilon} \quad \text{---} (3)$$

$\beta_0 = \frac{1}{c}\sqrt{\frac{2}{m_e}}$, $\Gamma_a = \frac{\hbar}{\tau_a}$ (τ_a is the lifetime of bound positron in the relation to annihilation). Taking $\varepsilon \sim E \sim 0.1$ ev,

10^{-10} sec, Z_b 10, we obtain from equation (3) $\frac{\sigma_a}{\sigma_D} \sim 10^3$ If E is neglected in (3) in comparison with ε , then the value of the level of ε may be determined from (3) on the basis of values of $\frac{\sigma_a}{\sigma_D}$ and τ_a determined by experiment. In the case of the molecules of CCl_4, for which σ_a/σ_D =702 (P-63) and $\tau_a = 2 \times 10^{-10}$ sec, we find in this manner from (3) $\varepsilon \approx 0.07$ ev (Z_b=32 was taken, as in (P-63). The premises itself concerning the existance of the bound of positron may be substantiated in the case of approximately spherical-symmetrical molecule by assuming that in these cases the interaction of positron with molecule is described by some central symmetric potential V(r). The value of the shallow level in the case of such a potential may be determined on the basis of considerations analogous to those uesed in the theory of slow particle scattering. Let the losely bound state be described by the equation

$$\frac{d^2y}{dr^2} + \left(-x^2 - u(r)\right) y = 0, \quad x^2 = \frac{2m_e E}{\hbar^2}, \quad U = \frac{2m_e V}{\hbar^2}$$

and let x be much smaller than the effective radius R of the potential. Then x^2 may be neglected in the region $r \ll 1/x$ in (4) and, consequently the solution of y is derived from the simplified equation $y_0'' - Uy_0 = 0$ having the asymptote $y_0 = C(1-A.r)$ where C is the normalising factor, A the constant dependent only on the properties of the potential $V(r)$. In the intermediate region $R \ll r \ll 1/x$ this asymptote should coincide with the asymptote of equation (4). In other words the level of ε is defined by the equation $x = a$. In the case of a potential of the type $V = -(r^2+R^2)^2$ for which the equation $y_0'' - Uy_0 = 0$ has a simple solution ($\alpha = pe^2 a_0 \cdot ^3$, a_0 is the Bohr radius, p is the polarizability), the depth of the level of ε found in the manner described is:

$$\varepsilon = \frac{\pi^2}{32} \frac{\hbar^2}{m_e R^2} \left(\sqrt{1+2p\left(\frac{a_0}{R}\right)^2} - 2\right)^2 \text{----} (5)$$

According to (5) the condition of the existance of the level is given by the inequality $p(a_0/R)^2 > 3/2$.
Assuming that R coincides with the gas kinetic radius of the particle approximately, we find that this inequality is not satisfied in the case of Ar (in accordance with the results in R-6) where it is shown that for Ar, σ_a/σ_D i.e. the resonance effect is absent in Ar.

For all other molecules investigated in (R-6) the inequality $p(a_0/R)^2 > 3/2$ exists, i.e the resonance effect for them exists. In case of CH_4 the value of the level found by formula (5) proves to be equal to $\varepsilon \approx 0.10$ ev. Taking for CH_4 this value of the level, the experimental value of $\sigma_a/\sigma_D = 20$ and $Z_b = 8$, we find from (3) the anihilation time τ_0 of the bound state in methane $\tau_a = 10^{-8}$ sec. In conclusion it should be noted that a test of the interpretation which has been set forth, would be observation of elastic scattering of slow positrons ($E \sim 0.1$ ev) in gases for which higher values of annihilation rates have been observed. In accordance with the foregoing the cross-sections σ_y of this process should considerably exceed the geometrical cross-sections of the molecules and be given by Wigner formula:

$$\sigma_y = \frac{\hbar^2}{2m_e} \Big/ \frac{1}{E+\varepsilon}$$

Appendix B

Phenomenological theory of annihilation from bound states.

(D.A.L.Paul)

For the purpose of this theory it is assumed that all positrons have slowed down to their ultimate velocity range,which is generally slightly above thermal, and that all positron formation and other higher energy phenomena are therefore complete. Four processes must be occuring, according to the Gol'danskii picture

(a) direct annihilation of free positrons at a rate λ
(b) formation of positron-molecule ions, rate λ_f
(c) dissociation of positron-molecule ions, rate λ_d
(d) annihilation of the bound positron, rate λ_b

The rates of change of the numbers of unbound and bound positrons will take place according to the usual differential laws. In this case

$$\dot{N} = -(\lambda + \lambda_f) + \lambda_d N_b$$

$$\dot{N}_b = -(\lambda_b + \lambda_d) + \lambda_f N$$

where N is the number of free positrons and N_b is the number of bound positrons.

Equations (1) and (2) have solutions of the form

$$N = A \exp(-\alpha t) + B(-\beta t)$$

$$N_b = C \exp(-\alpha t) + D(-\beta t)$$

where $\alpha, \beta = \lambda + \lambda_f + \lambda_b + \lambda_d \pm \sqrt{(\lambda + \lambda_f + \lambda_b + \lambda_d) + 4\lambda_f\lambda_d}$

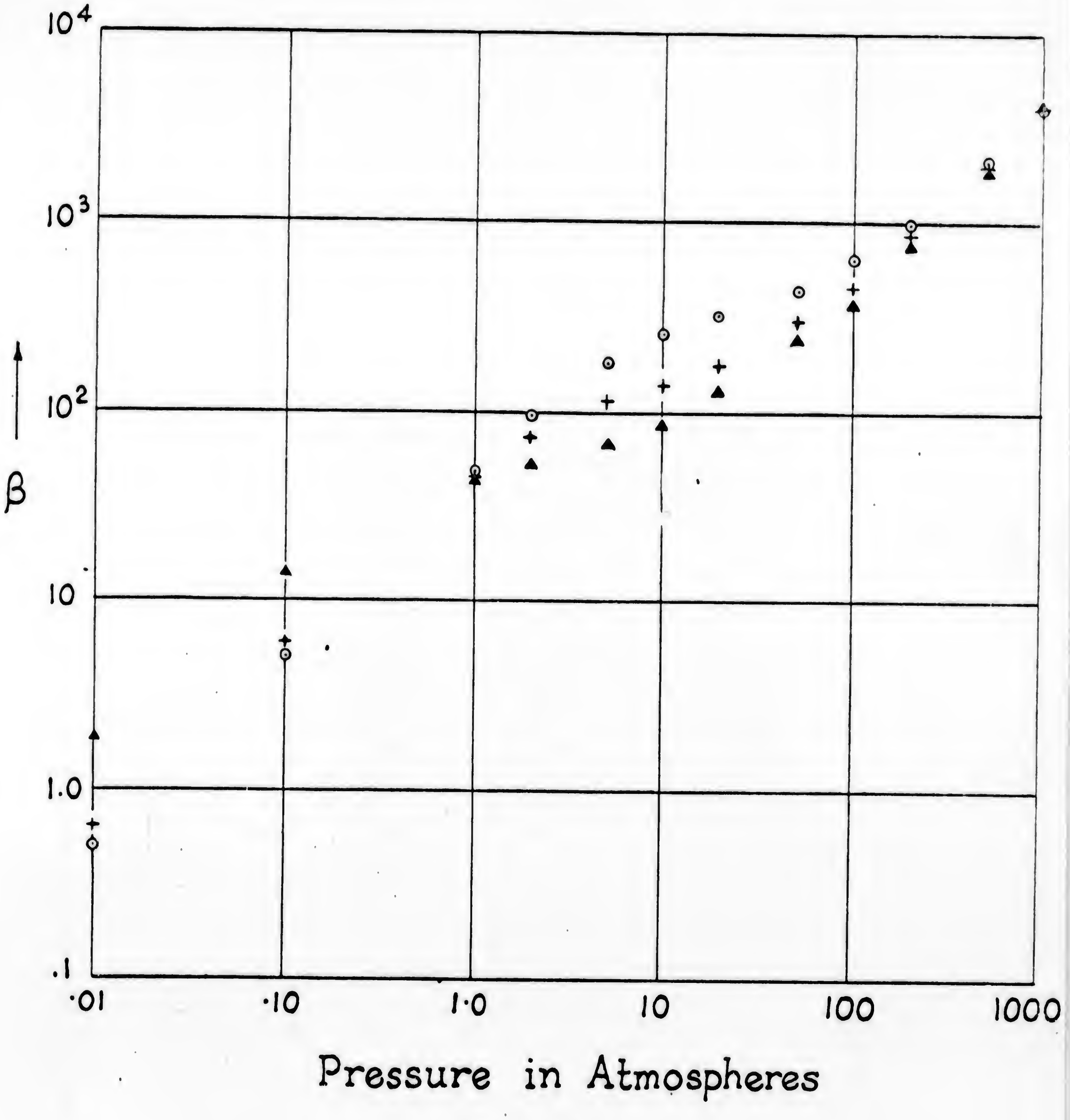

10^4
10^3
10^2
10
1.0
.1
β
·01
·10
1·0
10
100
1000
Pressure in Atmospheres

FIG. 12

Taking the positve square root to correspond to α, we note that the observed annihilation rate always corresponds to β. It is generally to be expected that λ and λ_f will be proportional to the pressure at low pressures, though λ_f may also have a significant quadratic pressure dependence at moderate and high pressures owing to many body collisions. The rate λ_p should be independent of pressure except perhaps at very high pressures (pressure broadening). The dissociation rate λ_d may be negligible if the "bound" state has an appreciable binding energy, and in particular is not a virtual state. If the binding energy is so small that molecular collisions can cause dissociation, λ_d may have a pressure dependence similar to λ. If the capturing state is really a virtual state, then λ_d will have a constant value in addition to any pressure dependence.

Initial conditions are taken to be:

$N_b = 0, \quad C + D \neq 0$

$$B/A = -\frac{\alpha - \lambda - \lambda_f}{\beta - \lambda - \lambda_f}$$

$A + B = N(0) = 1$ (in computation) i.e. for one positron. Therefore annihilation rate is equal to $\lambda N + \lambda_b N_b$. We have computed β for various values of the parameters. A log-log plot figure.12 of three sets of computed values of β against pressure in atmospheres has been made. To be in agreement with ~~exp~~ experiment at low pressures the slope of the log-log plot must be +1, and we have found that this is true for small dissociation rates $\lambda_d \ll \lambda_b$ In such cases this model agrees with Gol'danskii prediction, and is in keeping with experimental observation.

It is suggested that these measurements with methane may be continued at higher pressures.

References

(A-32) Anderson, C.D. Phys. Rev. 41 , 405 (1932).

(B-33) Blackett,P.M.S.,and Occhianlini,G.P.S.,
Proc. Roy. Soc. (London) A 139, 699(1933).

(D-30) Dirac, P.M.A., Proc. Camb. Phil. Soc. 26, 361 (1930).

(E-54) Endt, P.M.,and Kluyver, J.C., Rev. Mod. Phy. 26,95(1954).

(F-64) Falk, W.R., and Jones, Can. J. Phy 42, Sept.(1964).

(G-64) Gol'danskii,V.I.,and Sayasov,Yu.S. J.E.T.P. (1964).

(G-62) Graham, R.L., Geiger, R.E., and Barton, R.
Nuc. Instrm. Methods. 15, 360 (1962)

(H-61) Heymann, F.F., Osmon, P.E., Viet, J.J., and Williams,
W.F., Proc. Phys. Soc. (London) 78, 1o38 (1961).

(M-34) Mohorovicic, S., Astron. Nachr, 253, 94 (1934).

(P-58) Paul, D.A.L. Ph.D. thesis 1958.

(P-63) Paul, D.A.L., St. Pierre, L. Phys. Rev. Lett. 11 493
(1963).

(P-64) Paul, D.A.L., Proc. Phys. Soc. (London) 84, 563 (1964).

(R-45) Ruark, A.E., Phys. Rev. 68, 278 (1945).

(S-52) Smith, A.Lee and Johnston, H.L.,
J.Am. Chem. Soc.,74 , 4696 (1952).

(T-63) Tao, S.J., Green, J.H. and Celitans, G.J.
Proc. Phys. (London) 81, 1091 (1963).

(T-64) Tao, S.J., Bell, J. and Green, J.H.
Proc. Phys. Soc. (London) 83, 453 (1964).

(Y-50) Yang, C.N., Phys. Rev. 77, 242 (1950).